

# Multiple-scale integro-differential perturbation method for generic non-Markovian environments

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In order to approximate non-Markovian dynamics encountered in the theory of quantum open systems, a new multiple-scale perturbation method is generalized to integro-differential equations. Two (primary and auxiliary) time scales are particularly identified in the strong-coupling regime where the open system and its environment are strongly interacted with evident non-Markovianity being revealed. The new method provides acceptable approximation compared with traditional perturbation methods for generically structured reservoirs and indicates correct oscillatory and decay behaviors, helping us to recognize non-Markovianity in a perturbative way.

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## I. INTRODUCTION

It is unimaginable to study modern quantum dynamics without referring to the theory of quantum open systems nowadays. The quantum open system [1], as a statistical dynamic model, has induced tremendous new theoretical ideas and concepts such as dissipation and decoherence [2, 3], sudden death of entanglement [4, 5] and non-Markovianity [6–8] which are followed with increasing interest these days, and has directly or indirectly conducted experiments in many areas of quantum physics and quantum information, e.g., quantum computation and communication [9–11], quantum metrology [12], cavity quantum electrodynamics [13, 14], etc. It is understandable that to exactly solve the time-dependent evolution of a quantum open system must be of extreme difficulty for one has to handle infinite quanta with interaction between the system and environment, therefore perturbation approximation is required in most cases. Apparently, one of the modest perturbation methods is to use the ordinary differential perturbation (ODP) method [15] directly inasmuch as one can solve a series of Schrödinger differential equations in a complete Hilbert space of the entire closed system (the open system plus its environment). One can also consider the problem within the scenario of Liouville space and derive a master equation [16, 17] which describes the evolution of density operator of the open system using the *Nakajima-Zwanzig projection operator perturbation techniques* [18, 19]. The approximate result thus takes the form of a *generalized master equation* (GME) [19] or a *time-convolutionless master equation* (TCL) [20]. However, the range of applicability of these traditional perturbation methods is limited away from the *strong-coupling regime* where the system and environment are strongly coupled enough

that non-Markovianity is evidently revealed. This non-Markovian regime has exhibited many appealing features like bidirectional exchange of energy and/or coherence as well as singularity of evolution but has still remained not fully understood yet, partially because of the failure of the traditional perturbation methods above.

It seems not probable to find a useful perturbation method in the strong-coupling regime, since traditionally one only chooses the perturbation parameter to represent the global system-environment interaction strength and only allows it to be small. In this paper, however, we select the dimensionless perturbation parameter  $\alpha$  straight from the spectrum of the environment instead. Even if different types of spectrum result in completely different evolutions [21–23], the asymptotic behaviors therein are similar as the coupling strength (which  $\alpha$  is negatively correlated with) grows. In Section II, we limit the problem to two-level open systems and thus formulate the evolution as an integro-differential equation accordingly. As we shall see, the non-unitary and complex dynamics in the strong-coupling regime implies multiple (time) scales [24]. Hence, in Section III, we generalize a new multiple-scale integro-differential perturbation method for integro-differential equations; a primary time scale  $T$  and an auxiliary time scale  $\tau$  are specially introduced for the non-Markovian open-system problem. In Section IV, we compare the method with traditional perturbation methods, examine the non-Markovian behavior, and investigate the corresponding evolution within differently structured reservoirs. Finally, in Section V, the two-level problem is reconsidered in situations where the dipole-dipole interaction and more than one photon are involved. We expect that this multiple-scale perturbation method not only provides a new mathematical approximation approach but also contributes to a better knowledge of the relation between time scales and non-Markovianity in quantum mechanics [25].

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## II. TWO-LEVEL OPEN SYSTEM IN THE STRONG-COUPLING REGIME

To gain insight into the dynamics of a two-level open system, we begin by considering a Hamiltonian in the interaction picture [1]

$$H_I(t) = \sum_k \sum_{\omega_A} g_k(\omega_A) A^\dagger(\omega_A) E_k e^{i(\omega_A - \omega_k)t} + \text{h.c.} \quad (1)$$

which describes an open system  $A$  coupled to an environment  $E$  consisting of numerous discrete modes  $k$ .  $A(\omega_A)$  and  $A^\dagger(\omega_A)$  are annihilation and creation operators acting on  $A$  where  $\omega_A$  labels possible energy transitions. The operators  $E_k$  and  $E_k^\dagger$  labeled by mode  $k$  act on  $E$  with  $\omega_k$  the corresponding mode frequency.  $g_k(\omega_A)$  specifies the coupling strength. In the case of a two-level system coupled with a quantized radiation bath, the only transition is allowed by central frequency  $\omega_0$  between the two levels, and Eq. (1) is simplified to

$$H_I(t) = \hbar \sum_k g_k \sigma_+ \otimes a_k e^{i(\omega_0 - \omega_k)t} + \text{h.c.} \quad (2)$$

where  $a_k$  and  $a_k^\dagger$  are annihilation and creation operators in the radiation field and  $\sigma_\pm$  Pauli operators [1]. As the *rotating-wave approximation* [26] is already applied (i.e., the *ultra-strong coupling regime* [27] is unreachable by assuming  $\omega_0 \ll g_k$  when the frequency of mode  $k$  is near  $\omega_0$ ), the photon number remains conserved, and one can thus introduce independent Hilbert complete subspaces constructed by vectors with different photon numbers

$$\begin{aligned} &|g\rangle_A \otimes |0\rangle_E; \\ &|e\rangle_A \otimes |0\rangle_E, \quad |g\rangle_A \otimes |1_k\rangle_E; \\ &|e\rangle_A \otimes |1_j\rangle_E, \quad |g\rangle_A \otimes |1_j 1_k\rangle_E; \\ &\vdots \end{aligned}$$

$|g\rangle_A$  and  $|e\rangle_A$  are the ground and excited states of the two-level system, respectively. One aims to build independent sets of integro-differential equations from these subspaces by applying the Schrödinger equation to the entire closed system  $A \otimes E$  [1]. We further restrain the problem by handling at most *one* photon in the entire system, and the evolution of therein restrained wave function  $|\psi(t)\rangle = C_0(t)|g\rangle_A \otimes |0\rangle_E + C(t)|e\rangle_A \otimes |0\rangle_E + \sum_k D_k(t)|g\rangle_A \otimes |1_k\rangle_E$  is given by [26]

$$\begin{aligned} \dot{C}_0 &= 0, \quad \dot{C} = -i \sum_k g_k e^{i(\omega_0 - \omega_k)t} D_k, \\ \dot{D}_k &= -i g_k^* e^{-i(\omega_0 - \omega_k)t} C. \end{aligned}$$

With no difficulty a closed integro-differential equation for  $C$  can be derived,

$$\dot{C}(t) = - \int_0^t dt' G(t, t') C(t'). \quad (3)$$

The right-hand side of Eq. (3) is a convolution integral, where  $G(t, t')$  acts as a second-order correlation function as well as a non-Markovian memory kernel

$$\begin{aligned} G(t, t') &= \sum_k g_k g_k^* e^{i(\omega_0 - \omega_k)(t - t')} \\ &\simeq \int_0^\infty d\omega J(\omega) e^{i(\omega_0 - \omega)(t - t')} \end{aligned} \quad (4)$$

of which the Fourier transform is the *spectral density*  $J(\omega)$  of the environment [1]. Different shapes of  $J(\omega)$  characterize types of radiation bath with completely different behaviors [21]. However, there should be a universal dimensionless factor, defined as  $\alpha$ , distinguishing the bandwidth of  $J(\omega)$  relative to its central frequency. The spectral density is altered globally from a Dirac peak to a flat spectrum as  $\alpha$  goes from zero to infinity and shows complicated structure in the range. A neat example appears in the study of cavity-QED where we may choose  $\alpha \sim Q^{-1/2}$  with  $Q$  the quality factor of cavity [26]. Next, for the purpose of applying perturbation methods independently of dimension scales, we substitute time quantities in Eq. (3) by introducing a dimensionless quantity  $\tilde{t} = \gamma t$  where  $\gamma \sim J(\omega_0)$  describes the absolute coupling strength between system and environment. The new dimensionless correlation function  $\tilde{G}(\tilde{t}, \tilde{t}')$  takes a general form,

$$\tilde{G}(\tilde{t}, \tilde{t}') \sim \alpha^p G(\alpha^q(\tilde{t} - \tilde{t}')),$$

with integers  $p, q > 0$  accordingly.

The evolution of system  $A$  can also be written into the form of an exact quantum master equation [1],

$$\begin{aligned} \dot{\rho}_A(t) &= -\frac{i}{2} S(t) [\sigma_+ \sigma_-, \rho_A(t)] \\ &+ \Gamma(t) \left( \sigma_- \rho_A(t) \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho_A(t) \} \right), \end{aligned} \quad (5)$$

where  $\rho_A(t) = \text{Tr}_E \{ |\psi(t)\rangle \langle \psi(t)| \}$  is the reduced density operator (suppose ensemble  $A \otimes E$  is *not* mixed w.l.o.g.). The time-dependent coefficient  $S(t) = -2\Im \{ \dot{C}(t)/C(t) \}$  represents a Lamb-shift-like renormalized Hamiltonian and  $\Gamma(t) = -2\Re \{ \dot{C}(t)/C(t) \}$  represents a dissipator [1]. One can verify that the solution of the upper-level population of  $\rho_A(t)$  from Eq. (5) is  $|C(t)|^2$ , in agreement with Eq. (3).

It is of special interest to consider the difference of dynamics between  $\alpha \gg 1$  which gives the *weak-coupling regime* and  $\alpha \ll 1$  where the so-called *strong-coupling regime* locates [1]. In the weak-coupling regime, a traditional perturbation method tracing on the small factor  $\alpha^{-1}$  can always work well, no matter it is the original equations of quantum states or types of generalized quantum master equations [16, 18–20] that are used, or even the *Born-Markov approximation* [2] is enough to give a pure and simple result in the Markovian limit. On the other hand, we will see in Section IV that none of the perturbation methods are suitable in

the strong-coupling regime where complex evolution and strong non-Markovianity are involved.

### III. MULTIPLE-SCALE INTEGRO-DIFFERENTIAL PERTURBATION METHOD

It is never easy to find a good approximation for the evolution of Eq. (3). We know  $C(t)$  has to change its behavior from oscillation to pure exponential decay when  $\alpha$  goes from zero to infinity. The presence of both oscillatory and decay behaviors in the strong-coupling regime implies non-unitary and complex dynamics. Besides, one has to take care of the singularity of  $\tilde{G}(\tilde{t}, \tilde{t}')$  when  $\alpha \rightarrow 0$  and ask for  $|C(t)|$  to be physically smaller than  $|C(0)|$  all the time. Nevertheless, a typical perturbation method known as the *multiple-scale perturbation method* [24] can, more or less, handle similar problems in ordinary differential equations. The procedure of the multiple-scale perturbation method goes as follows: first, one chooses two or more different (time) scales and guesses their forms; second, he/she treats the scales as independent variables and transforms the original equations into a series of partial differential equations; finally, he/she needs to make sure higher perturbation terms diverge no more quickly than former terms [24]. Based on the same thought, we choose two possible time scales in our non-Markovian integro-differential equation: a primary time scale  $T$  and an auxiliary time scale  $\tau$  as our *ansatz*,

$$\begin{aligned} T &= (A_0\alpha + A_1\alpha^3 + \cdots + A_n\alpha^{2n+1} + \cdots) t, \\ \tau &= (B_0\alpha^2 + B_1\alpha^4 + \cdots + B_n\alpha^{2n+2} + \cdots) t, \end{aligned} \quad (6)$$

and also their dimensionless forms  $\tilde{\tau} = \gamma\tau$  and  $\tilde{T} = \gamma T$ . The coefficients  $\{A_n\}$  and  $\{B_n\}$  are undetermined yet. The differential operator acts locally and can thus be divided into two independent partial differential operators,

$$\frac{d}{dt} = \sum_n A_n \alpha^{2n+1} \frac{\partial}{\partial \tilde{T}} + \sum_n B_n \alpha^{2n+2} \frac{\partial}{\partial \tilde{\tau}}. \quad (7)$$

The integral operator, on the contrary, acts globally and cannot be separated independently. To apply the multiple-scale method to integro-differential equations, we therefore have to avoid dealing with integral terms directly. Here, the trick is to bring Eq. (3) into higher differential orders and try to cancel out integral terms one by one. We find the trick can succeed provided the correlation function  $\tilde{G}(\tilde{t}, \tilde{t}')$  is analytic at  $\tilde{t} = \tilde{t}'$ , i.e., its power expansion

$$\tilde{G}(\tilde{t}, \tilde{t}') \simeq G_0\alpha^p + G_1\alpha^{p+q}(\tilde{t} - \tilde{t}') + G_2\alpha^{p+2q}(\tilde{t} - \tilde{t}')^2 + \cdots \quad (8)$$

exists near  $\tilde{t} = \tilde{t}'$ .  $\{G_n\}$  are  $n$ th-derivative coefficients. Unfortunately, some well-studied structured reservoir such as PBG(photonic band-gap)-like medium [28] does

not satisfy the criteria of Eq. (8) but has singularity  $\sim (\tilde{t} - \tilde{t}')^{1/2}$ . Now it is assured by Eq. (8) that every perturbation integral terms in Eq. (3) will eventually be canceled out by derivatives owing to the cancellation formula  $(d/d\tilde{t})^{n+1} \int d\tilde{t}' (\tilde{t} - \tilde{t}')^n C(\tilde{t}') = n!C(\tilde{t})$ . Finally, expanding  $C(\tilde{t})$  into perturbation terms gives rise to the final form of Eq. (3),

$$\begin{aligned} & \left( \sum_n A_n \alpha^{2n+1} \frac{\partial}{\partial \tilde{T}} \right) \sum_n \alpha^n C^{(n)}(\tilde{T}, \tilde{\tau}) \\ & + \left( \sum_n B_n \alpha^{2n+2} \frac{\partial}{\partial \tilde{\tau}} \right) \sum_n \alpha^n C^{(n)}(\tilde{T}, \tilde{\tau}) \\ & = - \int_0^{\tilde{t}} d\tilde{t}' \sum_n G_n \alpha^{p+nq} (\tilde{t} - \tilde{t}')^n \sum_n \alpha^n C^{(n)}(\tilde{t}'). \end{aligned} \quad (9)$$

The iterating perturbation process with Eq. (9) as our initial equation contains two steps:

1. Let  $d/d\tilde{t}$  act on both sides of Eq. (9), cancel out a "global" integral term, then branch into two time scales in the form of Eq. (7).
2. Draw terms with the lowest perturbation order out of Eq. (9) as the corresponding perturbation equation pending to be solved.

As Eq. (9) becomes more and more "local" during iteration, the integral expansions in Eq. (9) will be canceled in ascending order provided  $p > 1$ . In other words, we can consider the perturbation order of  $\int d\tilde{t}' (\tilde{t} - \tilde{t}')^n$  as  $\alpha^{-n-1}$  "locally". The resulted equation given by the terms taken from the second step should merely be differential and therefore can be solved by regular multiple-scale methods [24].

### IV. EXAMPLES OF NON-MARKOVIAN ENVIRONMENTS

#### A. Lorentz reservoir

In this subsection, we introduce the *damped Jaynes-Cummings model* [1] as our first important example since it can be exactly solved by many different methods such as the Laplace transform or the pseudo/quasi-mode pictures [29]. This cavity-QED model describes the coupling between two-level system and field modes in a single cavity. The field modes are characterized by a Lorentzian spectral density [1]

$$J(\omega) = \frac{\gamma\lambda^2}{2\pi [(\omega_0 - \omega)^2 + \lambda^2]} \quad (10)$$

when the coupling between the system and reservoir is on resonance. Here,  $\lambda$  is the spectral width and  $\gamma$  the coupling strength. The Fourier transform of Eq. (10)

yields the correlation function

$$G(t, t') = \frac{1}{2} \gamma \lambda e^{-\lambda(t-t')}. \quad (11)$$

Substituting Eq. (11) into Eq. (3) one derives  $C(t) = e^{-\lambda t/2} [\cos(Dt/2) + (\lambda/D) \sin(Dt/2)]$  with initial conditions given by  $C(0) = 1$  and  $\dot{C}(0) = 0$  [1]. The parameter  $D = \sqrt{2\gamma\lambda - \lambda^2}$  distinguishes different coupling regimes: when  $\gamma < \lambda/2$ , the system and environment are weakly coupled, and  $C(t)$  evolves unidirectionally; when  $\gamma > \lambda/2$ ,  $D$  is real, and  $C(t)$  evolves with oscillation [1].

### 1. Comparison of perturbation methods

In order to demonstrate advantages of the new perturbation method, we compare it with traditional perturbation methods including the ODP, GME, and TCL methods. The latter two methods involve derivation of approximate master equations for the damped Jaynes-Cummings model (see Appendix A). Calculation results of the evolution of the upper-level population  $|C|^2$  by different traditional perturbation methods in the strong-coupling regime are shown in Fig. 1 with details of calculation presented in Appendix B.

Behaviors of the perturbation methods in the strong-coupling regime are complicated (see Fig. 1): we find that the secular ODP terms all diverge and cannot provide correct approximations; in spite of convergence of the results, the TCL method only reflects unidirectional decay behaviors of the system while loses its oscillatory characteristics; although the GME shows similar oscillatory behaviors, the oscillating frequency is mismatched and the population even takes negative values. It is also implied that the perturbation results of any higher order are not in agreement with the exact solution. Traditional perturbation methods in the strong-coupling regime therefore all fail.

It is essential to explain why these methods in the strong-coupling regime are not applicable. The singularity in Eq. (9) induces a small boundary layer where  $C(t)$  varies so fast when  $\alpha^p \rightarrow 0$  that any order of ODP solution cannot extend out of the layer and has to diverge [24]. From TCL master equations only analytical  $\Gamma_T(t)$  can be derived (see Eq. (B2)) which will never approach the exact  $\Gamma(t)$  in Eq. (5) which in this case is not only negative but even singular so as to exhibit its strong non-Markovianity. Finally, in addition to the disadvantage of losing positivity in Eq. (B1), the assumption in Eq. (A3) that  $\text{Tr}_A \{\rho(t)\} \approx \rho_E$  is time-independent in GME method is also invalid in the strong-coupling regime (so is TCL). In fact, the dependence of  $\rho_E(t)$  on time is strong enough, driven by the Rabi oscillation between the system and environment.

Next, we apply the multiple-scale integro-differential perturbation method to the damped Jaynes-Cummings model. Since different coupling regimes in the model are

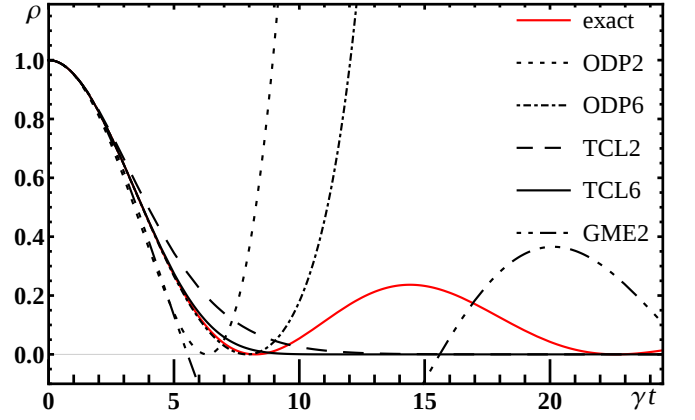


FIG. 1: (Color online) Approximate solutions of upper-level population in the strong-coupling regime of the damped Jaynes-Cummings model [1] from traditional perturbation methods [15, 18–20].  $|C(0)|^2 = 1$ ,  $\gamma = 1$ , and  $\lambda = 1/10$ .

simply distinguished by the width-strength ratio  $\lambda/\gamma$ , we make an attempt by choosing  $\alpha^p = \alpha^q = \alpha^2 = \lambda/\gamma$ , which put in Eq. (9) together with Eq. (11) gives  $G_0 = 1/2, G_1 = -1/2, G_2 = 1/4, \dots$ . Following the proposed two steps in Section III one can obtain a second-order partial differential equation for  $C^{(0)}(\tilde{T}, \tilde{\tau})$  to the perturbation order of  $\alpha^2$ ,

$$\alpha^2 A_0^2 \frac{\partial^2}{\partial \tilde{T}^2} C^{(0)} = -\alpha^2 G_0 C^{(0)}, \quad (12)$$

where the right-hand side is the largest perturbation term in  $(d/d\tilde{t}) \int d\tilde{t}' G_0 \sum_n \alpha^{2+2n} C^{(n)}(\tilde{t}')$ . Executing the two-step loop again yields a third-order differential equation to the order of  $\alpha^4$ ,

$$\begin{aligned} & \alpha^4 A_0^3 \frac{\partial^3}{\partial \tilde{T}^3} C^{(1)} + 2\alpha^4 A_0^2 B_0 \frac{\partial^3}{\partial \tilde{T}^2 \partial \tilde{\tau}} C^{(0)} \\ &= -\alpha^4 A_0 G_0 \frac{\partial}{\partial \tilde{T}} C^{(1)} - \alpha^4 G_1 C^{(0)}. \end{aligned} \quad (13)$$

The solution of Eq. (12) is

$$C^{(0)}(\tilde{T}, \tilde{\tau}) = E_0(\tilde{\tau}) \cos \frac{\tilde{T}}{\sqrt{2}A_0} + F_0(\tilde{\tau}) \sin \frac{\tilde{T}}{\sqrt{2}A_0}.$$

As  $C^{(1)}(\tilde{T}, \tilde{\tau})$  should not dominate the solution rather than  $C^{(0)}(\tilde{T}, \tilde{\tau})$  does, we must have the sum of  $C^{(0)}$  terms in Eq. (13) equal to zero so that  $C^{(1)}$  is bounded. Along with the initial conditions we find  $E_0(\tilde{\tau}) = e^{-\tilde{\tau}/(2B_0)}$  and  $F_0(\tilde{\tau}) = 0$ , as well as the solution of Eq. (13),

$$C^{(1)}(\tilde{T}, \tilde{\tau}) = E_1(\tilde{\tau}) \cos \frac{\tilde{T}}{\sqrt{2}A_0} + F_1(\tilde{\tau}) \sin \frac{\tilde{T}}{\sqrt{2}A_0}.$$

Higher-order differential equations indicate  $E_1(\tilde{\tau}) = 0$ ,  $F_1(\tilde{\tau}) = e^{-\tilde{\tau}/(2B_0)}/\sqrt{2}$ ,  $A_1/A_0 = -1/4$ , and  $B_1/B_0 = 0$

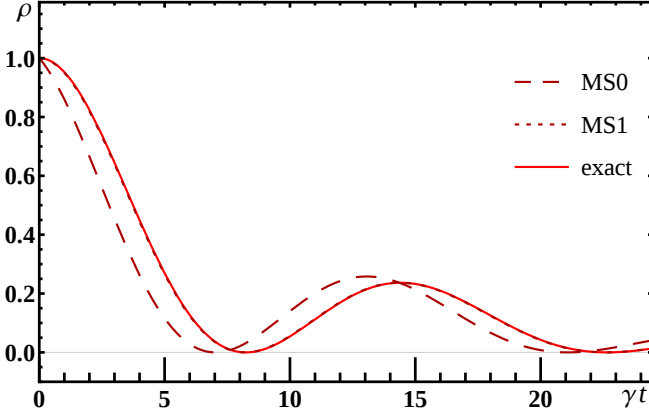


FIG. 2: (Color online) Approximate solutions of upper-level population in the strong-coupling regime of the damped Jaynes-Cummings model [1] from the multiple-scale integro-differential perturbation method.  $|C(0)|^2 = 1$ ,  $\gamma = 1$ , and  $\lambda = 1/10$ .

(see Appendix C). Figure 2 shows that the first-order perturbation solution (MS1)  $|C^{(0)} + \alpha C^{(1)}|^2$  is already in acceptable agreement with the exact solution and maintains positive and bounded.

## 2. Non-Markovianity

It is known that a quantum Markovian evolution defined by a set of trace-reserving linear maps  $\{\mathcal{E}(t, t'), t \geq t'\}$  where  $\mathcal{E}(t, t')$  is the propagator for the open system  $A$  defined by Eq. (A4) requires *complete positivity* for its propagators under the composition law [6]

$$\mathcal{E}(t, t'') = \mathcal{E}(t, t')\mathcal{E}(t', t''), \quad t \geq t' \geq t''.$$

Within the form of Eq. (5), the requirement gives rise to the *Gorini-Kossakowski-Sudarshan-Lindblad theorem*: a quantum evolution described by Eq. (5) is Markovian if and only if  $\Gamma(t) \geq 0$  for any time  $t$  [6]. Furthermore, the dissipator  $\Gamma(t)$  can be expressed in given time scales,

$$\Gamma(T, \tau) = -2\Re \left\{ \sum_n A_n \alpha^{2n+1} \frac{\partial}{\partial T} \ln C \right\} - 2\Re \left\{ \sum_n B_n \alpha^{2n+2} \frac{\partial}{\partial \tau} \ln C \right\}. \quad (14)$$

Apparently, sequences  $\{A_n\}$  and  $\{B_n\}$  determine the radius of convergence of the series around  $\alpha = 0$ . In this case of a Lorentzian reservoir, the sequence  $\{B_n\}$  terminates for  $B_1 = 0$ ; the second term in Eq. (14) simply

gives a positive constant  $-\gamma G_1/G_0$  with no contribution to the non-Markovianity. On the other hand, the partial dependence of  $C$  on the primary time scale  $T$  is not monotonic, so is  $\ln C$ . Hence, the evolution must be non-Markovian unless the first series in Eq. (14) diverges, making  $T$  trivial. In other words, the existence of the primary time scale is a signal for non-Markovianity. In addition, it is worth noting that by expanding the parameter  $D = \sqrt{2\gamma\lambda - \lambda^2}$  from the exact solution around  $\alpha = 0$ , one finds the coefficients in

$$D \simeq \sqrt{2\gamma} (\alpha - \alpha^3/4 + \dots)$$

coincide with  $\{A_n\}$ . It is very likely that the radius of convergence of  $\{A_n\}$  plays as the role of distinguishing the non-Markovian regime for generic reservoirs, though the derivation of  $\{A_n\}$  (and  $\{B_n\}$ ) proves to be somewhat complicated (see Appendix C).

Another noticeable non-Markovian feature related to time scales is the existence of minimal evolution time [30] between possible states and steep decrease [30] of the quantum (open-system) speed limit [31] in the strong-coupling regime. It was discovered that the evolution time between two arbitrarily possible pure/mixed states is not unique if the evolution is non-Markovian [30]. Here, clearly it is the existence of the primary time scale that speeds up the evolution. If we write down the master equations in the form of Eq. (5) given by

$$\Gamma_{\text{MS0}}(t) = \lambda + \sqrt{2\gamma\lambda} \tan \left[ \frac{\sqrt{2\gamma\lambda}}{2} t \right],$$

$$\Gamma_{\text{MS1}}(t) = \left\{ 4\gamma + 2\sqrt{2\gamma\lambda} \tan \left[ \frac{\sqrt{2\gamma\lambda}}{2} \left( 1 - \frac{\lambda}{4\gamma} \right) t \right] \right\}^{-1} \cdot \left\{ \lambda^2 + (4\gamma + \lambda) \sqrt{2\gamma\lambda} \tan \left[ \frac{\sqrt{2\gamma\lambda}}{2} \left( 1 - \frac{\lambda}{4\gamma} \right) t \right] \right\},$$

and  $S_{\text{MS0}}(t) = S_{\text{MS1}}(t) = 0$  corresponding to solutions MS0 and MS1, respectively, we can see that the dissipators have infinite numbers of singularities, among which their first singularities are at  $\hat{t}_{\text{MS0}} = \pi/\sqrt{2\gamma\lambda}$  and  $\hat{t}_{\text{MS1}} = (\gamma\lambda/2)^{-1/2} (1 - \lambda/4\gamma)^{-1} \cos^{-1} \sqrt{\lambda/(2\gamma + \lambda)}$ . In fact, all the singularities correspond to infinite possibilities of evolution time between two orthogonal states (with the maximal distance), and  $\hat{t}_{\text{MS0}}$  ( $\hat{t}_{\text{MS1}}$ ) is the minimal evolution time defined therein [30]. Compared with the exact solution  $\hat{t} = (2/D)[\pi - \tan^{-1}(D/\lambda)]$ , relative errors of the minimal evolution time for two orthogonal states in MS0 and MS1 are to the order of  $\alpha$  and  $\alpha^3$ , respectively. We note that the minimal evolution time cannot be identified by traditional perturbation methods, either.

## B. Generic reservoirs

In this subsection, we apply the multiple-scale integro-differential method to other generic reservoirs with

similar definitions of  $\lambda$  and  $\gamma$  like the Lorentzian spectral density in Eq. (10) and investigate how the results are related to the specific form of  $G(t, t')$ . The perturbation

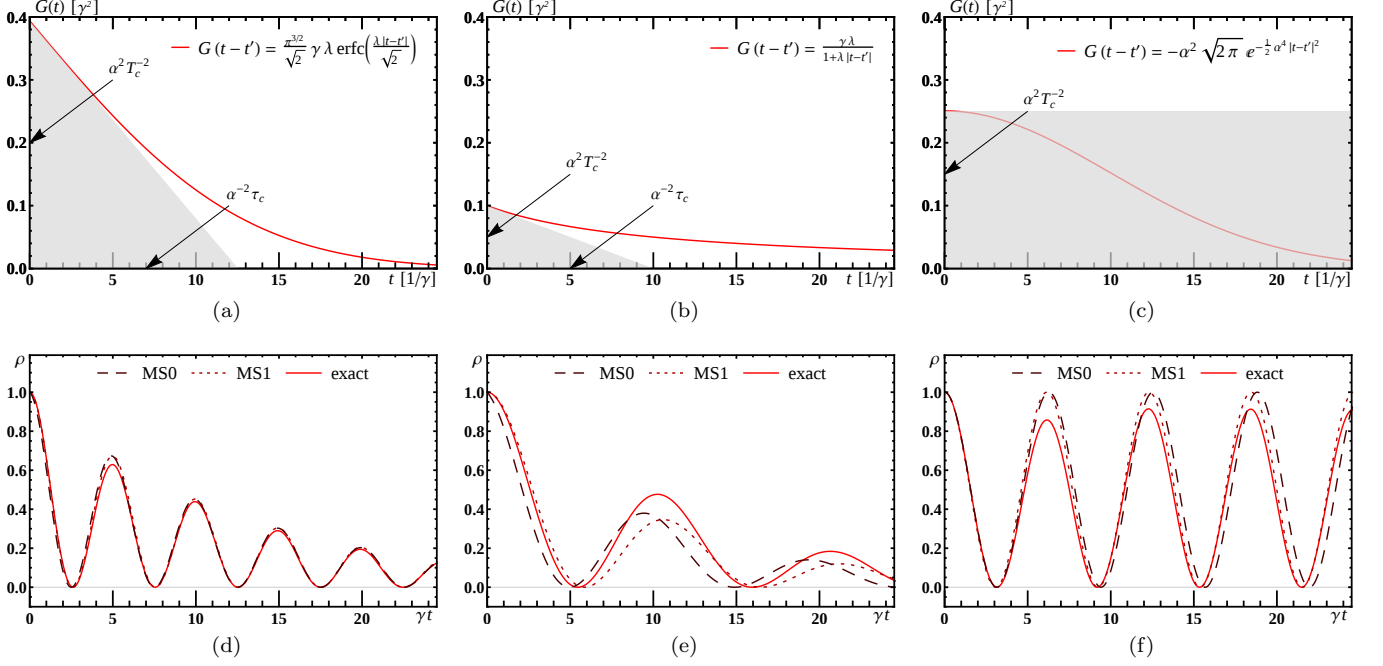


FIG. 3: (Color online) Generic reservoirs characterized by second-order correlation functions in the form of (a) a Gaussian error distribution, (b) an inverse-law distribution, and (c) a Gaussian distribution, with their corresponding approximate solutions of upper-level population from the multiple-scale integro-differential perturbation method given in (d), (e), and (f), separately.  $|C(0)|^2 = 1$ ,  $\gamma = 1$ , and  $\lambda = 1/10$ .

solutions should also take neat forms like those of the damped Jaynes-Cummings model with separated time scales as variables if we still choose  $\alpha \sim (\lambda/\gamma)^{1/2}$ . The characteristic time for the two primary and auxiliary time scales  $T_c$  and  $\tau_c$  are related to the first two coefficients of the series of  $G(t, t')$  by  $T_c \sim \alpha G_0^{-1/2}$  and  $\tau_c \sim \alpha^2 G_1$ , respectively, which indicates that the global behavior of  $C(t)$  is determined locally at  $t \sim t'$  resulted by the successful analytic expansion of  $G(t, t')$  in Eq. (8). One would expect that the oscillatory and decay behaviors in the strong-coupling regime are sufficiently contributed by the two time scales distinctly like they do in Lorentz reservoir, but we will see this is not the case.

In Fig. 3, three different correlation functions are chosen with corresponding perturbation solutions demonstrated and compared. The first reservoir characterized by a Gaussian error function results in acceptable approximation compared with the exact numeric solution (see Fig. 3(d)). As expected,  $\operatorname{erfc}(x) \sim o(x^{-1} \exp(-x^2))$  descends fast enough to make Eq. (8) more "locally" dominant. In comparison to the first one, the second reservoir characterized by an inversed-law correlation function results in worse approximation (see Fig. 3(e)), because not only  $o(x^{-1})$  descends much slower which causes Eq. (8) to be less "local", but in fact the non-integrable inverse-law correlation function leads to log-singularity of the corresponding spectral density  $J(\omega)$  at  $\omega \sim \omega_0$  as well. It is rather remarkable that the third reservoir characterized by a Gaussian function results

in completely different approximation (see Fig. 3(f)) from the previous two examples. Since this smooth and symmetric  $G(t, t')$  gives rise to  $G_1 = 0$ , the auxiliary time scale  $\tau$  collapses to infinitesimal and becomes trivial. However, as there is only one attractor at  $C(\infty) = 0$  since  $J(\omega)$  has wings extended to infinity [21], the global decay still exists and must be contained in the other time scale(s). The pure oscillating solution of  $T$  implies that our choice of the time scales in Eq. (6) does not apply to this example and the correct time scales should be more complicatedly entangled, or even non-linear.

## V. BEYOND SIMPLE TWO-LEVEL OPEN SYSTEM

The multiple-scale integro-differential perturbation method can naturally be fitted into any types of generalized problems as long as non-Markovian evolutions therein can be written into a set of integro-differential equations with memory kernels well defined. We shortly discuss its possible generalization in this section.

*Dipole-dipole interaction.*—Duplicated systems under dipole-dipole interaction are a typical example which *ad lib* fits into our method, since the entire Hamiltonians simply include  $N$  individual system Hamiltonians interacting with a shared reservoir in the form of Eq. (1) and

$N(N-1)/2$  dipole-dipole Hamiltonians in the form

$$H_{ij} = \hbar K_{ij} (\sigma_+^i \sigma_-^j + \sigma_-^i \sigma_+^j)$$

which also keeps photons conserved [32]. The complete subspaces are thus constructed by

$$\begin{aligned} & \left\{ \prod_i^N |g\rangle_{A_i} \right\} \otimes |0\rangle_E; \\ & \left\{ |e\rangle_{A_n} \otimes \prod_{i \neq n}^N |g\rangle_{A_i} \right\} \otimes |0\rangle_E, \quad \left\{ \prod_i^N |g\rangle_{A_i} \right\} \otimes |1_k\rangle_E; \\ & \vdots \end{aligned}$$

The perturbation procedure is almost exactly the same, except the order of the dipole-dipole coupling strength coefficients  $K_{ij}$  requires to be initially defined by  $\tilde{K}_{ij} = \alpha^s K_{ij}$ . Different order  $s$  identifies different *dipole-dipole coupling sub-regimes* and influences non-Markovianity as well as speed of disentanglements between  $N$  systems directly [32]. One will anticipate that the hardest part of calculation is to derive  $N$ -order determinants which arise in the orthogonalization of linearly coupled differential perturbation equations.

*Beyond one photon.*—It still remains a problem how to solve an open system with more than one photon involved in its reservoir, because the only knowledge  $J(\omega)$  we know about the environment is not enough to determine the entire evolutions. For example, in the *two-photon* subspace we consider how the wave function  $|\psi(t)\rangle = \sum_j C_j(t) |e\rangle_A \otimes |1_j\rangle_E + \sum_{\{j,k\}} D_{jk}(t) |g\rangle_A \otimes |1_j 1_k\rangle_E$  should evolve. The Schrödinger equation gives rise to (noticing  $D_{jk} = D_{kj}$ )

$$\begin{aligned} \dot{C}_j &= -i \sum_k g_k e^{i(\omega_0 - \omega_k)t} D_{jk}, \\ \dot{D}_{jk} &= -i g_j^* e^{-i(\omega_0 - \omega_j)t} C_k - i(1 - \delta_{jk}) g_k^* e^{-i(\omega_0 - \omega_k)t} C_j, \end{aligned}$$

from which the infinite numbers of formatted integro-differential equations contain  $g_j^* g_k$  cross-terms and cannot be expressed with  $J(\omega)$ . An approach which can more or less address the problem is to re-discretize  $J(\omega)$  into  $g_k$  ( $g_k^*$ ) terms and only consider the first finite  $N_k$  modes [21]. Hence, the approximate evolutions shrink into  $N_k$  coupling integro-differential equations where our method can thus be tried. We note that the same discretization procedure can also be generalized for  $n$ -level systems (e.g., the two-band quantum-dot model [33, 34]).

## VI. CONCLUSION

Firstly, we present a new multiple-scale integro-differential perturbation method that can be mathematically applied to some kinds of integro-differential equations. The particular application of the multiple-scale method to non-Markovian two-level quantum open

systems indicates that there should be two time scales—a primary time scale  $T$  and an auxiliary time scale  $\tau$ —both dominating the non-Markovian evolution in the strong-coupling regime where the system and its environment are strongly coupled. It is revealed that none of the traditional perturbation methods [15, 18–20] works in the non-Markovian regime. The failures include secular divergent terms, absence of oscillatory characteristics, and invalid negative populations. Instead, the multiple-scale method can provide good approximation for generic reservoirs, provided the autocorrelation function  $G(t, t')$  descends fast enough to be “local” so that the auxiliary time scale will not collapse.

Identification of negative population/coherence flow in the master equation in terms of  $T$  and  $\tau$ , in addition, implies that the existence (convergence) of the primary time scale should be a signal for non-Markovianity. The oscillatory and decay behaviors in the strong-coupling regime are separately contributed by the two time scales in most cases, yet there exist counterexamples (e.g., the Gaussian reservoir). Last but not least, singularities of evolution as well as the minimal evolution time [30] in the non-Markovian regime can also be identified by the multiple-scale integro-differential perturbation method, with relative error following the perturbation order. In the future, we expect our method to be of assistance to specific time-related issues in the quantum open system theory; meanwhile we look forward to expanding the potential of our work to somewhat general topics, e.g., multiple-scaling behaviors, complex networks, etc.

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## Appendix A: Perturbation Methods on Derivation of Master Equations

*Generalized master equation (GME).*—An ordinary differential perturbation (ODP) method can only work for a group of differential equations given by the Schrödinger equation directly. In practice, it is necessary to use a density operator in Liouville space instead of state vectors to quantify statistically mixed quantum states; the evolution of the density operator for system  $A$  should be trace-reserving under any approximation. To this end, one introduces the density operator  $\rho(t)$  for  $A \otimes E$  (suppose  $\rho(t) = |\psi(t)\rangle\langle\psi(t)|$  a *pure* state w.l.o.g.) and two Nakajima-Zwanzig projection super-operators  $\mathcal{P}$  and  $\mathcal{Q}$ :  $\mathcal{P}\rho(t) = \mathcal{P}^n \rho(t) = \text{Tr}_E \{\rho(t)\} \otimes \text{Tr}_A \{\rho(t)\}$  and



$\mathcal{Q} = 1 - \mathcal{P}$  [18, 19]. It can be proved from the Liouville equation

$$\dot{\rho}(t) = -\frac{i}{\hbar} [H_I(t), \rho(t)] = \alpha^{-1} \mathcal{L}(t) \rho(t) \quad (\text{A1})$$

that  $\mathcal{P}\mathcal{L}(t)\mathcal{L}(t_2)\cdots\mathcal{L}(t_{2n+1})\mathcal{P} = 0$  [1]. Functional iteration techniques [15] yield the following equation [1],

$$\begin{aligned} \mathcal{P}\dot{\rho}(t) &= \alpha^{-1} \mathcal{P}\mathcal{L}(t) \mathcal{G}(t, 0) \mathcal{Q}\rho(0) \\ &+ \alpha^{-2} \mathcal{P} \int_0^t dt' \mathcal{L}(t) \mathcal{G}(t, t') \mathcal{Q}\mathcal{L}(t') \mathcal{P}\rho(t'), \end{aligned} \quad (\text{A2})$$

where  $\mathcal{G}(t, t') = \mathcal{T}_{\leftarrow} \exp \left[ \alpha^{-1} \int_{t'}^t dt'' \mathcal{Q}\mathcal{L}(t'') \right]$ . Perturbation process can be applied after unfolding  $\mathcal{P}\rho(t) = \sum_{n=0} \alpha^{-n} \mathcal{P}\rho^{(n)}(t)$ . If  $\mathcal{Q}\rho(0) = 0$ , only the second term of Eq. (A2) exists. Since  $\mathcal{L}(t)$  and  $\mathcal{P}\rho(t)$  are all separated, one can consider the system and the environment separately and calculate the terms  $\mathcal{L}(t)\mathcal{L}(t')\cdots\mathcal{L}(t^{(2n)})\mathcal{P}\rho(t)$  in Eq. (A2). The large degree of freedom of the environment guarantees  $\text{Tr}_A \{\rho(t)\} \approx \rho_E$  in statistical equilibrium if the coupling between the system and the environment is weak ( $\alpha^{-1} \ll 1$ ) [1]. One then has

$$\text{Tr} \left\{ \sum_k E_k E_k^\dagger \rho_E \right\} = \text{Tr} \left\{ \sum_k E_k^\dagger E_k \rho_E \right\} + 1 = 1 \quad (\text{A3})$$

taken into Eq. (A2) as there is only one photon considered in the reservoir.

*Time-convolutionless (TCL) master equation.*—The integral in Eq. (A2) keeps non-Markovian behaviors retained in the GME at the expense of simpler solutions. As a tradeoff, one can eliminate the convolution integral while maintaining some time-dependent behaviors using the time-convolutionless master equations [20]. One notices from Eq. (A1) that  $\rho(t') = \mathcal{E}(t', t) (\mathcal{P} + \mathcal{Q}) \rho(t)$  where the propagation operator is

$$\mathcal{E}(t', t) = \mathcal{T}_{\rightarrow} \exp \left[ -\alpha^{-1} \int_{t'}^t dt'' \mathcal{L}(t'') \right]. \quad (\text{A4})$$

Taking it into Eq. (A2) gives

$$\mathcal{P}\dot{\rho}(t) = \mathcal{K}(t) \mathcal{P}\rho(t) + \mathcal{I}(t) \mathcal{Q}\rho(0), \quad (\text{A5})$$

with super-operators  $\mathcal{K}(t) = \alpha^{-1} \mathcal{P}\mathcal{L}(t) [1 - \Sigma(t)]^{-1} \mathcal{P}$  and  $\mathcal{I}(t) = \alpha^{-1} \mathcal{P}\mathcal{L}(t) [1 - \Sigma(t)]^{-1} \mathcal{G}(t, 0) \mathcal{Q}$ , in which  $\Sigma(t) = \alpha^{-1} \int_0^t dt' \mathcal{G}(t, t') \mathcal{Q}\mathcal{L}(t') \mathcal{P}\mathcal{E}(t', t)$  [1]. One then derives the TCL master equations by unfolding  $[1 - \Sigma(t)]^{-1} = \sum_{n=0} [\Sigma(t)]^n$ . The loss of ability to characterize non-Markovianity in the TCL method results from the elimination of convolution integrals as manifested in Section IV.

## Appendix B: Approximate Results of the Lorentzian Reservoir from Traditional Perturbation Methods

*Ordinary differential perturbation.*—With an exponential correlation function (see Eq. (11)), one can directly construct a second-order differential equation from Eq. (3), which along with initial conditions  $C(0) = 1, \dot{C}(0) = 0$  generates the following perturbation solutions:

$$\begin{aligned} C^{(0)}(\tilde{t}) &= 1; \\ C^{(2)}(\tilde{t}) &= -\frac{1}{4} \tilde{t}^2; \\ C^{(4)}(\tilde{t}) &= \frac{1}{96} (8\tilde{t}^3 + \tilde{t}^4); \\ C^{(6)}(\tilde{t}) &= -\frac{1}{5760} (120\tilde{t}^4 + 24\tilde{t}^5 + \tilde{t}^6); \end{aligned}$$

... The results plotted in Fig. 1 are

$$\begin{aligned} \text{ODP-2:} \quad & \left( C^{(0)} + \alpha^2 C^{(2)} \right)^2; \\ \text{ODP-6:} \quad & \left( C^{(0)} + \alpha^2 C^{(2)} + \alpha^4 C^{(4)} + \alpha^6 C^{(6)} \right)^2. \end{aligned}$$

*Generalized master equation.*—The GME to the second order approximates the damped Jaynes-Cummings model by [1]

$$\begin{aligned} \dot{\rho}_A(t) &= \gamma \lambda \int_0^t dt' e^{-\lambda(t-t')} \\ & \left( \sigma_- \rho_A(t') \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho_A(t') \} \right). \end{aligned} \quad (\text{B1})$$

Solution of the upper-level population

$$\text{GME-2:} \quad e^{-\lambda t/2} \left[ \cos \left( \frac{D_G t}{2} \right) + \frac{\lambda}{D_G} \sin \left( \frac{D_G t}{2} \right) \right]$$

(with  $D_G = \sqrt{4\gamma\lambda - \lambda^2}$ ) is plotted in Fig. 1.

*Time-convolutionless master equation.*—The TCL master equation of the damped Jaynes-Cummings model takes a similar form of Eq. (5) [1],

$$\dot{\rho}_A(t) = \Gamma_T(t) \left( \sigma_- \rho_A(t) \sigma_+ - \frac{1}{2} \{ \sigma_+ \sigma_-, \rho_A(t) \} \right), \quad (\text{B2})$$

where  $\Gamma_T(t) = \Gamma_T^{(2)}(t) + \Gamma_T^{(4)}(t) + \Gamma_T^{(6)}(t) + \cdots$ . Here, the  $\alpha^{2n}$ -order term calculated from Eq. (A5) is complicated [1]:

$$\begin{aligned} \Gamma_T^{(2n)}(t) &= 2(-1)^{n+1} \int_0^t dt_1 \int_0^{t_1} dt_2 \cdots \int_0^{t_{2(n-1)}} dt_{2n-1} \\ & \langle f(t-t_1) f(t_2-t_3) \cdots f(t_{2n-2}t_{2n-1}) \rangle_{\text{oc}}, \end{aligned}$$

where the tedious expansion rule of the so-called *ordered cumulants* term  $\langle \cdots \rangle_{\text{oc}}$  is given in Ref. [1]. We only show



the first several terms here,

$$\begin{aligned}\Gamma_T^{(2)}(t) &= \gamma (1 - e^{-\lambda t}); \\ \Gamma_T^{(4)}(t) &= \frac{\gamma^2}{2\lambda} (1 - 2\lambda t e^{-\lambda t} - e^{-2\lambda t}); \\ \Gamma_T^{(6)}(t) &= \frac{\gamma^3}{4\lambda^2} (2 + e^{-\lambda t} - 2\lambda t e^{-\lambda t} - 2\lambda^2 t^2 e^{-\lambda t} \\ &\quad - 2e^{-2\lambda t} - 4\lambda t e^{-2\lambda t} - e^{-3\lambda t});\end{aligned}$$

... The results of the upper-level population plotted in Fig. 1 are

$$\text{TCL-2: } \exp \left[ - \int_0^t dt_1 \Gamma_T^{(2)}(t_1) \right];$$

$$\text{TCL-6: } \exp \left[ - \int_0^t dt_1 \left( \Gamma_T^{(2)}(t_1) + \Gamma_T^{(4)}(t_1) + \Gamma_T^{(6)}(t_1) \right) \right].$$

### Appendix C: Identify $\{A_n\}$ and $\{B_n\}$ in Multiple-Scale Perturbation Process

In the case of  $p = q = 2$ , similar to Eqs. (12) and (13), one is able to continue obtaining perturbation equations to higher orders. From Eqs. (C1) and (C2) (to the order of  $\alpha^6$  and  $\alpha^8$ ) one takes out  $C^{(0)}$  terms to find

$$A_1 = \left( \frac{3}{8} \frac{G_1^2}{G_0^3} - \frac{G_2}{G_0^2} \right) A_0$$

and

$$B_1 = \left( -\frac{G_1^2}{G_0^3} + 4\frac{G_2}{G_0^2} - 6\frac{G_3}{G_1 G_0} \right) B_0,$$

respectively.

$$\begin{aligned}& A_0^4 \frac{\partial^4}{\partial \tilde{T}^4} C^{(2)} + 3A_0^3 B_0 \frac{\partial^4}{\partial \tilde{T}^3 \partial \tilde{\tau}} C^{(1)} + 3A_0^2 B_0^2 \frac{\partial^4}{\partial \tilde{T}^2 \partial \tilde{\tau}^2} C^{(0)} + 2A_0^3 A_1 \frac{\partial^4}{\partial \tilde{T}^4} C^{(0)} \\ &= -A_0 B_0 G_0 \frac{\partial^2}{\partial \tilde{T} \partial \tilde{\tau}} C^{(1)} - A_0^2 G_0 \frac{\partial^2}{\partial \tilde{T}^2} C^{(2)} - A_0 G_1 \frac{\partial}{\partial \tilde{T}} C^{(1)} - 2G_2 C^{(0)}.\end{aligned}\tag{C1}$$

$$\begin{aligned}& A_0^5 \frac{\partial^5}{\partial \tilde{T}^5} C^{(3)} + 4A_0^4 B_0 \frac{\partial^5}{\partial \tilde{T}^4 \partial \tilde{\tau}} C^{(2)} + 6A_0^3 B_0^2 \frac{\partial^5}{\partial \tilde{T}^3 \partial \tilde{\tau}^2} C^{(1)} + 4A_0^2 B_0^3 \frac{\partial^5}{\partial \tilde{T}^2 \partial \tilde{\tau}^3} C^{(0)} \\ &+ 3A_0^4 A_1 \frac{\partial^5}{\partial \tilde{T}^5} C^{(1)} + 2A_0^4 B_1 \frac{\partial^5}{\partial \tilde{T}^4 \partial \tilde{\tau}} C^{(0)} + 8A_0^3 A_1 \frac{\partial^5}{\partial \tilde{T}^4 \partial \tilde{\tau}} C^{(0)} \\ &= -A_0 B_0^2 G_0 \frac{\partial^3}{\partial \tilde{T} \partial \tilde{\tau}^2} C^{(1)} - A_0^2 A_1 G_0 \frac{\partial^3}{\partial \tilde{T}^3} C^{(1)} - 2A_0^2 B_0 G_0 \frac{\partial^3}{\partial \tilde{T}^2 \partial \tilde{\tau}} C^{(2)} - A_0^3 G_0 \frac{\partial^3}{\partial \tilde{T}^3} C^{(3)} \\ &\quad - A_0 B_0 G_1 \frac{\partial^2}{\partial \tilde{T} \partial \tilde{\tau}} C^{(1)} - A_0^2 G_1 \frac{\partial^2}{\partial \tilde{T}^2} C^{(2)} - 2A_0 G_2 \frac{\partial}{\partial \tilde{T}} C^{(1)} - 6G_3 C^{(0)}.\end{aligned}\tag{C2}$$

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